

ポリアルキルメタクリレート (PAMA) 系高分子添加剤の分子量および極性が 境界潤滑性能に及ぼす影響

Effects of molecular weight and polarity of polyalkyl methacrylate (PAMA) polymer additives on boundary lubrication performance

名大 (院)・工 (学) *ソン ユシ, 名大 (院)・工 (非) 張 俊彦, 名大・工 (正) 福澤 健二,

名大・工 (兼) JST さきがけ (正) 伊藤 伸太郎,

名大・工 (兼) JST ACT-X (正) 東 直輝, 名大・情 (正) 張 賀東

Song Yuxi¹, Zhang Junyan¹, Kenji Fukuzawa¹, Shintaro Itoh^{1,2}, Naoki Azuma^{1,3}, Hedong Zhang¹

¹Nagoya University, ²JST PRESTO, ³JST ACT X

1. Introduction

To improve the energy efficiency, the viscosity of lubricant oils has been decreased. Nevertheless, the use of low-viscosity oils has limitations in creating sufficient dynamic pressure to fully separate sliding surfaces in fluid lubrication. This issue has led to a rise in the number of lubrication systems operating in mixed and boundary lubrication regimes, resulting in increased friction and wear. To reduce the wear and friction in mixed and boundary lubrication regimes, various friction modifiers have been added into the lubricant oil. Polymer additives such as polyalkylmethacrylate (PAMA) were found to be able to adsorb on sliding surfaces. The adsorbed films could separate the surfaces and thus protect the surfaces from direct contact. It was believed that the more additives adsorbed on surfaces, better friction and wear reduction performance could be achieved. However, the previous study on the organic friction modifiers (OFM) additives found that the adsorption mass or the thickness of adsorbed films only influences the friction coefficient during the first sliding rotation in the boundary lubrication regime¹⁾. As the sliding rotation increased, the correlation between adsorption mass and friction coefficient became negligible.

In this study, the thickness of adsorbed PAMA polymer film with different polarities and molecular weights was in-situ measured from lubricant oil with vertical-objective-type ellipsometric microscopy (VEM). After the thickness of adsorbed films saturated, the sliding experiments were conducted. The friction force during sliding under different loads were measured. After each sliding, the film thickness was measured and compared with the thickness before sliding. It was found the molecular weight did not influence the friction coefficient much. On the other hand, the friction coefficient was strongly affected by the polarity of the polymers. In addition, the strong correlation between adsorbed film thickness after sliding and friction coefficient was found.

2. Experiment methods

The schematic setup for measuring thickness of adsorbed film thickness with VEM is shown in Fig. 1²⁾. This method is based on ellipsometry. By analyzing the intensity image of the reflected polarized light captured by the CMOS camera, the gap between the glass substrate and the slider can be obtained. To measure the thickness of the adsorbed film, the slider was once separated from the glass substrate, allowing the polymer additive to adsorb onto the solid surfaces. After maintaining the separation for a period (~ 10 s), the slider then was firmly pressed on the glass substrate. The gap between the slider and the glass substrate was then measured by VEM while the slider was pressed. Since the base oil between the gap of slider and glass substrate was squeezed out during the pressing procedure, only the adsorbed film remains in the gap when the slider was pressed. Therefore, the measured gap at this moment is equal to the thickness of the adsorbed film. By repeating the procedures of separation and pressing, the temporal change in thickness of adsorbed film can be obtained.

In addition to film thickness measurement, the sliding mechanism was also added to measure the friction force during sliding, as shown in Fig. 2. In the setup, the slider was mounted on the biaxial flat springs, which consist of a stainless-steel flat spring that displaces vertically and a stainless-steel double-cantilever spring that displaces horizontally. When the glass substrate was moved horizontally by the x-piezo stage, the double-cantilever spring was bent due to the friction force acting on the slider. By measuring the displacement of the double-cantilever spring with the optical displacement sensor, the friction force

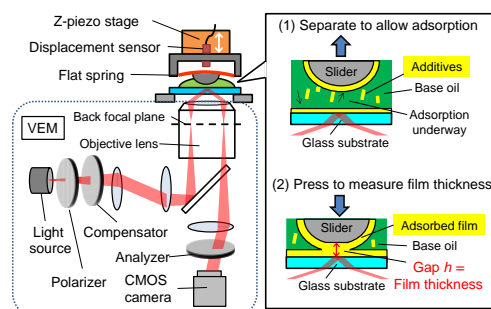


Fig. 1 Schematic setup for thickness measurement of adsorbed film with VEM

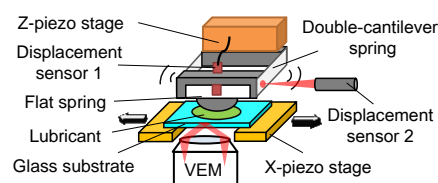


Fig. 2 Schematic setup for friction force measurement

could be obtained.

3. Experimental samples

The base oil used in this study is the group III mineral oil. The PAMA polymer was synthesized by mixing four methacrylate monomers: methyl methacrylate (C1), 2-ethylhexyl methacrylate (EH), lauryl methacrylate (LA) and stearyl methacrylate (ST). Among these monomers, C1 has the highest polarity, followed by EH, LA, and ST. To investigate the effects of polarity on adsorption and friction properties, three different PAMA polymers were synthesized by changing the concentration of C1 during copolymerization. These three PAMAs are PAMA-C1-5 % (C1: 5 wt%), PAMA-C1-10 % (C1: 10 wt%), and PAMA-C1-18 % (C1: 18 wt%). With the increase of C1 concentration, the polarity of PAMA also increases. PAMA polymers with molecular weights of ~20,000 (20 k) g/mol and ~60,000 (60 k) g/mol were used in this study. The concentration of the PAMA polymer additive is 2.0 wt%.

The glass with the refractive index of 1.93 was used as the substrate, and its thickness is 0.8 mm. The slider is the plano-convex glass lens, which was coated by the 53-nm thick stainless-steel through sputtering. Its radius is 15.6 mm. The roughness in Rq of the slider surface and the glass substrate was 1.1 nm and 0.4 nm, respectively.

4. Experimental procedure

The temporal change in thickness of adsorbed films of different polymers during adsorption in the lubricant oil was measured first. After the thickness of the adsorbed film was saturated, the glass substrate slid reciprocatingly at a constant speed driven by the x-piezo stage under different loads for 30 s. The amplitude of the triangular-wave sliding motion was set at 15 μ m. The sliding speed was 0.6 mm/s by setting the reciprocating frequency to 10 Hz. During each sliding movement, the friction force was measured. After each sliding movement, the film thicknesses were measured, and they were compared with the film thicknesses that before sliding. The loads during sliding were controlled by the z-piezo stage.

5. Results

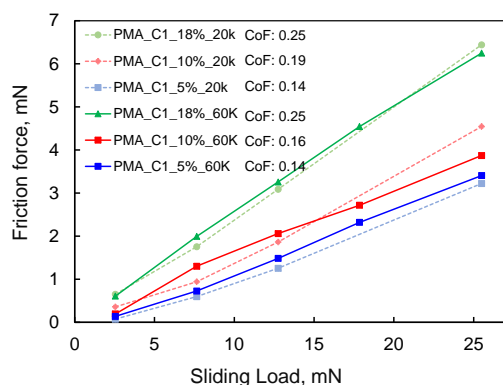


Fig. 3 Friction forces of polymer with different polarities and molecular weights

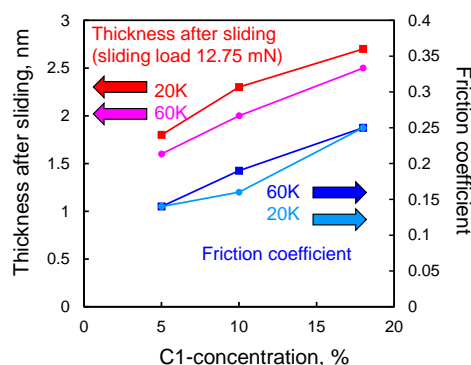


Fig. 4 Relationship between friction forces and adsorbed film thickness after sliding

Figure 3 shows measured friction force with different polymer additives. When the polarity of the polymer is the same, the difference of friction forces between polymer with the molecular weights of 20 k and 60 k was negligible. On the other hand, the measured coefficient of friction (CoF) increased as the polarity of polymer increases. Before sliding, the saturation thickness of the adsorbed for PAMA Mw 60 k with different polarities were similar. After sliding, the adsorbed film thickness of polymer with Mw 60 k decreased to the same level as that of Mw 20 k, as shown in Fig. 4. In addition, the strong correlation between friction coefficient during sliding and the adsorbed film thickness was found, as shown in Fig. 4. The thicker the adsorbed film after sliding, the higher the friction coefficient during sliding.

6. Summary

Thickness of adsorbed PAMA polymer films with different polarities and molecular weights were measured in-situ before and after sliding in lubricant oil. The correlation between friction coefficient and film thickness after sliding was found.

Acknowledgments

This study was partially supported by KAKENHI (20H00214) and ENEOS Corporation.

References

- 1) Fry, Benjamin M., et al. "Adsorption of organic friction modifier additives." *Langmuir* 36.5 (2020): 1147-1155.
- 2) Song, Yuxi, et al. "In-situ measurement of temporal changes in thickness of polymer adsorbed films from lubricant oil by vertical-objective-based ellipsometric microscopy." *Tribology International* 165 (2022): 107341.